IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:
Yoshimura et. al.
Serial No.: 10/594,627 Art Unit: 1796
Filed : July 16, 2007 Examiner: SERGENT, RABON A
Title : MEDICAL ADHESIVE

DECLARATION UNDER RULE 132

Honorable Commissioner of Patents and Trademarks,
Alexandria, Virginia 22313-1450

Sir:

I, Tetsuji Yoshimura, a citizen of Japan and having postal mailing address of 4-7-13, Kitayamadai, Konan-shi, Shiga 520-3241 JAPAN, declare and say that:

March 1979, I graduated from Graduate School of Science, Kyoto University, and received a master's degree in chemistry;

From April 1979, up to the present, I have been employed by Sanyo Chemical Industries, Ltd., and engaged in the works of research and development for organic polymers, especially in the field of polyurethane;

In June 2003, I joined the group where the inventors of the present invention belonged;

I am the inventor of the above-identified application and am familiar with the technical field of the present invention;

I have read the Official Action mailed and the references cited therein. I respectfully submit herewith my exact report;

In order to demonstrate the effect of a polyester polyol (B1-2) as the polyol component (B), monophenolic radical scavenger and a bisphenolic radical scavenger as a phenolic radical scavenger, and a different type of the fluorine-containing nonaromatic polyisocyanate component, I have carried out the following experiments.

<Production Example 7>

After 350 parts of ethylene glycol and 790 parts of adipic acid were fed into an autoclave and were dissolved at 120°C, the temperature inside the autoclave was elevated to 160°C while blowing nitrogen into a liquid phase and agitating. The reaction was continued for 4 hours while carrying out dehydration at normal pressure. The temperature inside the autoclave was further elevated to 230°C, and the reaction was continued for 25 hours while carrying out dehydration at normal pressure. The temperature inside the autoclave was cooled to 120°C. After 0.5 parts of sodium hydroxide were fed into an autoclave followed by substitution with nitrogen (the oxygen concentration in the vapor phase was 450 ppm), the inside of the autoclave was vacuum dehydrated for 60 minutes.

Then, after the mixture of 968 parts of ethylene oxide and 128 parts of propylene oxide was fed at 130°C for about 15 hours, the reaction was continued at 130°C for 3 hours, and liquid crude polyester polyol in which the content of oxyethylene groups was about 60% was obtained.

This liquid crude polyester polyol was treated with the synthetic magnesium silicate in the same method as that of the Production Example 1 and a purified polyester polyol (b7) was obtained. As for this (b7), the number

average molecular weight was 4,500, the content of oxyethylene groups was 60%, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Production Example 8>

After 620 parts of ethylene glycol, 490 parts of maleic acid anhydride and 1 part of phosphoric acid were fed into an autoclave followed by elevating the temperature inside the autoclave to 70°C while blowing nitrogen into a liquid phase and agitating, the reaction was continued for 2 hours. The temperature inside the autoclave was elevated to 140°C, and the reaction was continued for 7 hours. After 2 parts of sodium hydroxide were fed into an autoclave followed by substitution with nitrogen (the oxygen concentration in the vapor phase was 450 ppm), water and unreacted ethylene glycol were removed under vacuum for 8 hours.

Then, after 460 parts of ethylene oxide was fed at 170°C for about 5 hours, the reaction was continued at 170°C for 3 hours, and liquid crude polyester polyol in which the content of oxyethylene groups was about 55% was obtained.

This liquid crude polyester polyol was treated with the synthetic magnesium silicate in the same method as that of the Production Example 1 and a purified polyester polyol (b8) was obtained. As for this (b8), the number average molecular weight was 525, the content of oxyethylene groups was 55%, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Production Example 9>

After 760 parts of propylene glycol, 490 parts of maleic acid anhydride and 1 part of phosphoric acid were

fed into an autoclave, the temperature inside the autoclave was elevated to 70°C while blowing nitrogen into a liquid phase and agitating. The reaction was continued for 2 hours. The temperature inside the autoclave was elevated to 140°C, and the reaction was continued for 7 hours. After 2 parts of sodium hydroxide were fed into an autoclave followed by substitution with nitrogen (the oxygen concentration in the vapor phase was 450 ppm), water and unreacted ethylene glycol were removed under vacuum for 8 hours.

Then, after 606 parts of propylene oxide was fed at 170°C for about 5 hours, the reaction was continued at 170°C for 3 hours, and liquid crude polyester polyol in which the content of oxyethylene groups was about 55% was obtained.

This liquid crude polyester polyol was treated with the synthetic magnesium silicate in the same method as that of the Production Example 1 and a purified polyester polyol (b9) was obtained. As for this (b9), the number average molecular weight was 620, the content of oxyethylene groups was 0%, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Example 10>

The medical adhesive (P10) of the present invention was obtained by the same method as that in Example 1, except using 100 parts of the purified polyester polyol (b7) obtained in Production Example 7 as the polyol component (B) and 13.9 parts of bis(isocyanatomethyl)perfluorobutane (the ratio of NCO group/OH group = 2/1) as the fluorine-containing nonaromatic polyisocyanate component (A).

As for this (P10), the content of isocyanate groups

was 1.6%, the number average molecular weight (Mn) was 6,500, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Example 11>

The medical adhesive (P11) of the present invention was obtained by the same method as that in Example 1, except using 100 parts of the purified polyester polyol (b8) obtained in Production Example 8 as the polyol component (B) and 118.9 parts of bis(isocyanatomethyl)perfluorobutane (the ratio of NCO group/OH group = 2/1) as the fluorine-containing nonaromatic polyisocyanate component (A).

As for this (P11), the content of isocyanate groups was 7.3%, the number average molecular weight (Mn) was 750, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Example 12>

The medical adhesive (P12) of the present invention was obtained by the same method as that in Example 1, except using the mixture of 90 parts of the purified polyester polyol (b7) obtained in Production Example 7 and 10 parts of the purified polyester polyol (b9) obtained in Production Example 9 as the polyol component (B), and 22.5 parts of bis(isocyanatomethyl)perfluorobutane (the ratio of NCO group/OH group = 2/1) as the fluorine-containing nonaromatic polyisocyanate component (A).

As for this (P12), the content of isocyanate groups was 2.5%, the number average molecular weight (Mn) was 6,200, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Example 13>

The medical adhesive (P13) of the present invention was obtained by the same method as that in Example 12, except adding 0.5 parts of 2,6-di-t-butyl-4-ethylphenol (NOCTIZER M-17, manufactured by Ouchi Shinko Chemical Industrial Co., Ltd.) as the phenolic radical scavenger (PRS) in place of tetrakis-[methylene-3-(3',5'-di-t-butyl-4'-hydroxyphenyl)propionate]methane.

As for this (P13), the content of isocyanate groups was 2.4%, the number average molecular weight (Mn) was 6,100, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Example 14>

The medical adhesive (P14) of the present invention was obtained by the same method as that in Example 12, except adding 0.5 parts of 2,2'-methylenebis(4-ethyl-6-t-butylphenol) (Antage W-400, manufactured by Kawaguchi Chemical Industry Co., Ltd.) as the phenolic radical scavenger (PRS) in place of tetrakis-[methylene-3-(3',5'-di-t-butyl-4'-hydroxyphenyl)propionate]methane.

As for this (P14), the content of isocyanate groups was 2.5%, the number average molecular weight (Mn) was 6,200, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

<Example 15>

The medical adhesive (P15) of the present invention was obtained by the same method as that in Example 12, except using 29.7 parts of bis(isocyanatomethyl)perfluorohexane (the ratio of NCO group/OH group = 2/1) as the fluorine-containing

isocyanate component in place of 22.5 parts of bis(isocyanatomethyl)perfluorobutane.

As for this (P15), the content of isocyanate groups was 2.3%, the number average molecular weight (Mn) was 6,300, and the content of alkaline metals and/or alkaline earth metals was 0.01 mmol/kg.

Results

The medical adhesives (P10) to (P15) were evaluated by the procedures employed in Evaluations 1 and 2 of the specification of the present application. The results are shown in Table 1.

Table 1

	Example					
	10	11	12	13	14	15
Change in appearance	Excellent	Excellent	Excellent	Excellent	Excellent	Excellent
Wet adhesive strength (2H)	1.2	1.1	1.4	1.4	1.4	1.4
Wet adhesive strength (5D)	1.3	1.2	1.4	1.2	1.3	1.4

Conclusion

From the results shown above, the following facts 1 to 3 can be clearly read.

1. When the polyether polyol (B1-1) used in Example 1 is replaced by the polyester polyol (B1-2) (Example 10 to 12) respectively, Change in appearance, Wet adhesive strength (2H), and Wet adhesive strength (5D) are comparable before and after substitution. The effect of the present invention can be exerted.

2. When the polymer phenolic radical scavenger used in

Example 12 employing the polyester polyol (B1-2) is replaced by the monophenolic radical scavenger (Example 13) and the bisphenolic radical scavenger (Example 14) respectively, Change in appearance, Wet adhesive strength (2H), and Wet adhesive strength (5D) are comparable before and after substitution. The effect of the present invention can be exerted.

3. When the fluorine-containing isocyanate used in Example 12 employing the polyester polyol (B1-2) is replaced by the different type of fluorine-containing isocyanate (Example 15), Change in appearance, Wet adhesive strength (2H), and Wet adhesive strength (5D) are comparable before and after substitution. The effect of the present invention can be exerted.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Signed this day of , 2009

Tetsuji Yoshimura